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AN IMPROVED TECHNIQUE FOR THE DETERMINATION OF SPONTANEOUS COMBUSTION SUSCEPTIBILITY OF COALS

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by

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ABSTRACT

This report introduces a curve fitting technique based on cubic spline formulation to represent data obtained from crossing point experiments used for spontaneous combustion tests. The data are one dimensional in the form of temperature as a function of time. Using this formulation, the mathematical expression for the change and rate of change of temperature with respect to time are given. The proposed technique is used to fit data for two types of coal, high and low volatile. Spline coefficients, the change and the rate of change of temperature with time for these coals as determined using the technique are also included.

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DÉTERMINATION DE LA SUSCEPTIBILITÉ DES CHARBONS À LA COMBUSTION SPONTANÉE À L'AIDE D'UNE TECHNIQUE PERFECTIONNÉE

par

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RÉSUMÉ

Le présent rapport décrit une technique d'ajustement de courbe basée sur une formule spline cubique pour représenter les données obtenues lors des essais de point de passage réalisés au cours des essais sur la combustion spontanée. Les données obtenues sont d'une dimension, soit sous forme de température en tant que fonction de temps. Les expressions mathématiques représentant le changement et le taux de changement de température par rapport au temps sont déterminées à l'aide de cette formule. La technique proposée est utilisée pour ajuster les données obtenues lors des essais réalisés avec des charbons à haute et à faible teneur en matières volatiles. Les coefficients de spline, le changement et le taux le changement de température avec le temps déterminés pour ces charbons à l'aide de la technique susmentionnée sont aussi présentés dans le rapport.

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INTRODUCTION

It is well known that different types of coal have different spontaneous heating characteristics. With respect to its susceptibility to spontaneous combustion, these coals are categorized using a number of approaches. Most of these methods are based on two principles. The first principle makes use of petrological classification, or rank correlation but have not been found to be of enough practical use in distinguishing the liability of coals to spontaneous combustion. The latter approach is mainly based on measuring the oxygen avidity of coal, or on taking a record of thermal changes incurred in the process of oxidation. Two well established techniques have evolved on the basis of the second approach namely, crossing point and oxygen absorption. In the crossing point method, data obtained are in the form of temperature as a function of time. To obtain susceptibility index few other factors such as the change and the rate of change of temperature with time are important. Presently these factors are calculated manually. For greater accuracy in the analysis of data it is considered necessary to develop a semi empirical formula based on cubic spline to represent the experimental data obtained from spontaneous combustion tests. The following tasks are expected to achieve this objective:

1. develop a mathematical tool to represent crossing point data and to evaluate associated parameters required for spontaneous combustion susceptibilty index calculation.

2. automate existing spontaneous combustion system to obtain discrete data points to be used as an input to the mathematical model.

4. establish a spontaneous combustion index which is expected to be a function of temperature, the change and the rate of change with respect to time. The index may include other parameters if found useful to improve accuracy.

CROSSING POINT TEMPERATURE METHOD: BACKGROUND

This method is based on heating coal samples in an oxidizing atmosphere at a desired and programmed rate of temperature rise. Due to exothermicity of oxidation reaction, at some point the coal bed temperature exceeds the bath temperature which is commonly termed as

crossing point temperature. Highly susceptible coals to spontaneous heating have lower values of crossing point temperatures and the poorly susceptible coals have comparatively higher values. Various experimental techniques and designs of apparatus have been used in the past to determine the crossing point temperature of coals (1,2).

Over the years a considerable amount of work have been done in developing and improving the experimental technique for the determination of the crossing point temperature of coal. Unfortunately, there is very little recorded evidence in trying to explain or interpret the data obtained from these tests. In an earlier paper (3) by Feng, Chakravorty and Cochrane, some attempts have been made to develop an empirical index. This index was found to be useful in categorizing coals to some extent.

At the coal Reseach Laboratory, Canada Centre for Mineral and Energy Technology, (CANMET), the crossing point tests are carried out as follows. Coal samples in a stainless steel container are placed in a forced air oven where the temperature could be programmed and increased at a predetermined rate. The temperature of the coal sample and of the oven are measured on a continuous basis using two sets of thermocouples. The temperature of the coal is monitored and recorded as the temperature of the oven is increased in a controlled manner as discussed in an earlier report (4).

The samples are prepared using a standard procedure with the aim of reducing atmospheric oxidation to a minimum. Fresh coal samples immediately after sampling at the mine site is kept within a container in inert atmosphere. Coal samples are later crushed in a pulverizer also maintaining an inert atmosphere. Samples are then seived to -60 + 200 mesh size. All coal samples are moisture equilibriated at 60% R.H. over a 48 hour period to simulate actual atmospheric humidity conditions.

Air saturated at room temperature is passed through the coal bed at a rate of 80 ml/min and the temperature of the oven is increased at a rate of 1.0°C/min. upto 80°C and 0.5°C/min from 80°C to 210°C. Temperature of the oven and coal bed is monitored on a continuous basis during the experiment. Amount of coal sample used for each test is 50 gm.

MATHEMATICAL FORMULATIONS

The natural cubic spline, the first derivatives of which are continuous, was shown to best approximate the curve joining data points with minimum curvature. The set of functional values (temperature) $T_1,\ldots,T_M$ and the corresponding coordinates (time) $t_1, t_2,\ldots,t_M$, are fed into a natural cubic spline subroutine and an output array c of dimension (3,(L-1)) is obtained. The spline interpolate in the L-interval, (t_L-t_{L+1}), can then be constructed by

$$T_L(t) = \sum_{m=1}^{4} \alpha_m(t_L)\ (t-t_L)^{m-1} \qquad (1)$$

More than three data points are needed for the evaluation of the spline coefficients. The approximation accuracy increases with the number of given data points. The coefficients in a given interval, which are related to the first three derivatives of the approximation function at the interval boundaries, depend on all data points. Thus the effect of nonsystematic errors of individual data points on the numerical derivatives is inherently reduced resulting in "smooth derivatives" (4) (this property is partially attributable to the best approximation characteristic, and partially to the minimum curvature property of the cubic spline). Unlike a Taylor series approximation, the question of convergence is irrelevant here. Since all spline coefficients are equally important, none of them can be neglected without seriously reducing the approximation accuracy.

Since the first and second derivatives of the spline function are continuous, it is possible to determine the first and second derivative of Eq. (1).

Using Eq. (1), change and rate of change of temperature with respect to time can be determined as follows

$$\frac{\partial\, T_L(t)}{\partial\, t} = \sum_{m=2}^{4} \alpha_m(t_L)\ (m-1)(t-t_L)^{m-2}$$

$$= \alpha_2(t_L) + 2\alpha_3(t_L)(t-t_L) + 3\alpha_4(t-t_L)^2 \qquad (2)$$

and

$$\frac{\partial^2 T_L(t)}{\partial t^2} = \sum_{m=3}^{4} \alpha_m(t_L)\,(m-1)(m-2)(t-t_L)^{m-3}$$

$$= 2\alpha_3(t_L) + 6\alpha_4(t_L)(t-t_L) \qquad (3)$$

The spline coefficients α_1 ,α_2 ,α_3 and α_4 are unique for each interval.

CUBIC SPLINE AND CROSSING POINT EXPERIMENTS

Graphs obtained from crossing point tests have a general form similar to that of Fig. 1 (4). Results obtained from crossing point experiments for two different types of coal (low and high volatile bituminous coals) are shown in Fig. 1. In order to use the cubic spline to represent and to interpolate between data points, let us consider the curves for the high volatile coal A and the low volatile coal B as shown in Fig. 1. These curves can be represented by eight data points as given in Fig. 2 for coal A. The values for the time in minutes (X-axis) and temperature in °C (Y-axis) for the seven data points are then fed into a cubic spline routine. For the eight data points, there will be seven intervals (L=1,2,3,4,5,6,7) as shown in Fig. 2 for coal A. The routine calculates three coefficients for each interval, (α(L,1),α(L,2) and α(L,3)), which are then used to interpolate between the end points to calculate temperature, change and rate of change in temperature using Eqs.1-3. These calculations are given in Table 1 for the high volatile coal A. This table gives the measured and the corresponding interpolated temperature, calculated change and rate of change of temperature with time. It may be noted that in this case, the routine calculates the exact value of temperature (Table 1) as was fed into it. The calculated slope to the temperature-time curve (change of temperature with time) is positive at the selected temperatures and the curve characteristics can be explained as follows. The nature of the thermal curve depends on three distinct phenomena namely: 1) evaporation of moisture, 2) condensation of moisture, and 3) oxidation. Evaporation of moisture from coal results in an endothermic reaction, while the other two are exothermic. This curve is the product of all the concurrent reactions incured during the early stage of heating. It may be noted that the slope decreases for coal A from 0.53 at 81 °C to 0.39 at 96°C (mostly due to the endothermic reaction caused by evaporation of

moisture) and then increases to 0.525 at 107°C. As the temperature of the bath increases the slope increases more rapidly to 1.139 at 157°C. Similar trend is also observed for the low volatile bituminous coal B as shown in table 3. Results indicate that the slope first decreases from 0.56 at 93°C to 0.53 at 110°C, then the slope starts to increase to 0.598 at 142°C. It is important to note that the rate of increase of slope is slower in case of coal B in comparison to that of coal A. It is believed that the continuous increase in slope beyond 110°C is mainly due to lesser evaporation and progressively higher degree of oxidation.

Table 2 lists the spline coefficients in the six intervals. At each interval, the coefficients $\alpha(L,1)$ and $\alpha(L,2)$ are directly related to the first and second derivative of temperature with respect to time. Similar coefficients to those given in table 2 are given in table 4 for the low volatile coal B.

Visual examination of the temperature-time graphs for the two coals clearly shows that their nature are dependent upon the type of coal used. In particular, the change and the rate of change of temperature with time appeared to be unique for each coal. This is clearly indicated by the results given in tables 1 and 3. The rate of change of temperature with time for the high volatile bituminous coal A is significantly higher compared to that of the low volatile bituminous coal B. Further studies are in progress on a number of coals from different ranks so that this method can be used to characterize coals with respect to their liability to spontaneous combustion more effectively. It is obvious from the discussion made earlier that the spontaneous combustion susceptibility index, Sk, will have the following form:

$$Sk = f\ (T, DTM, DDTM)$$

where,

T = coal temperature
DTM = change of temperature with time
DDTM = rate of change of temperature with time.

It is intended to develop an empirical formula using this function to accurately represent the susceptibility index for different types of coal.

It is also proposed to carry out a set of experiments by passing saturated nitrogen through the coal bed and determine the effect of

moisture evaporation independent of oxidation on the spontaneous combustion characteristics of coal. This might demand a different interpretation technique based on evaluation of the area under the heating curves from both air and nitrogen experiments.

CONCLUSIONS

Cubic spline mathematical formulations appear to be a useful tool in representing crossing point data more effectively for categorizing coals with respect to their susceptibility to spontaneous combustion. Based on this formulation it may be possible to develop a susceptibility index which will indicate spontaneous combustion liability more accurately.

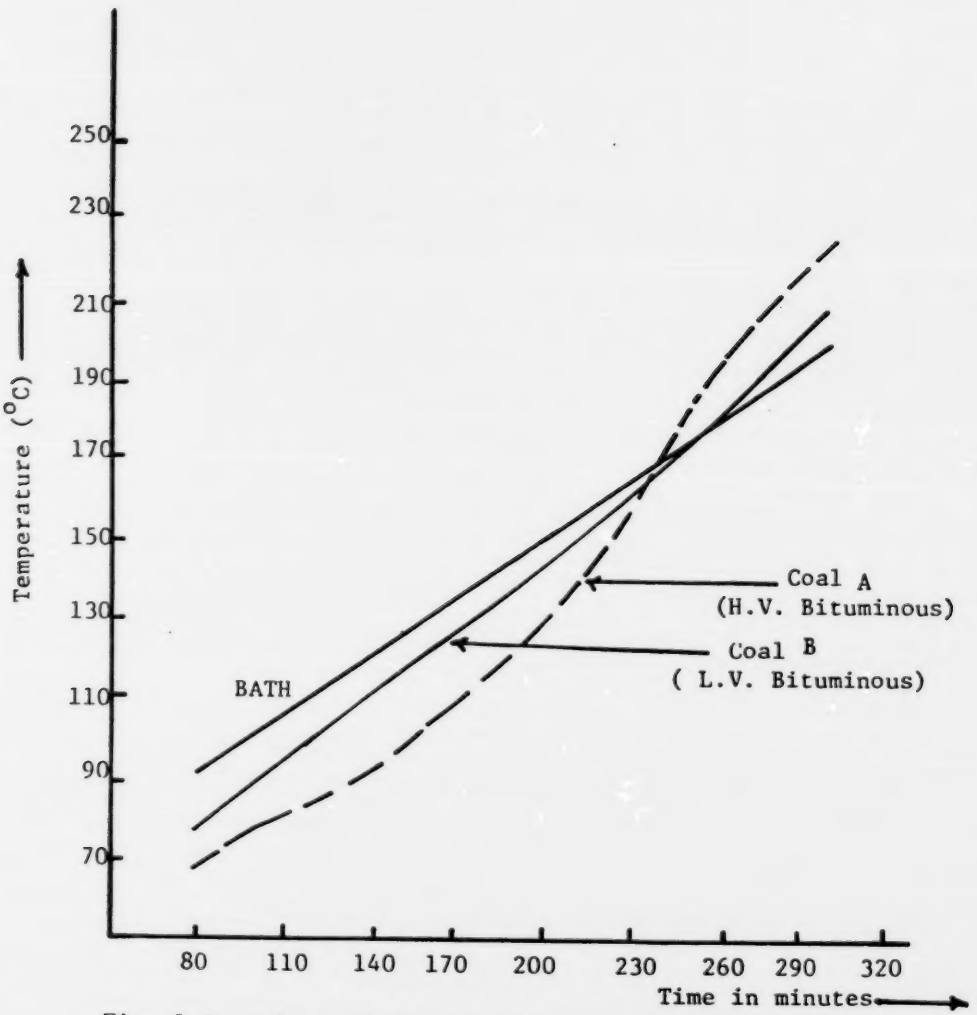

Fig. 1 Crossing point temperature graphs for two types of coals (High Volatile and low Volatile Bituminous).

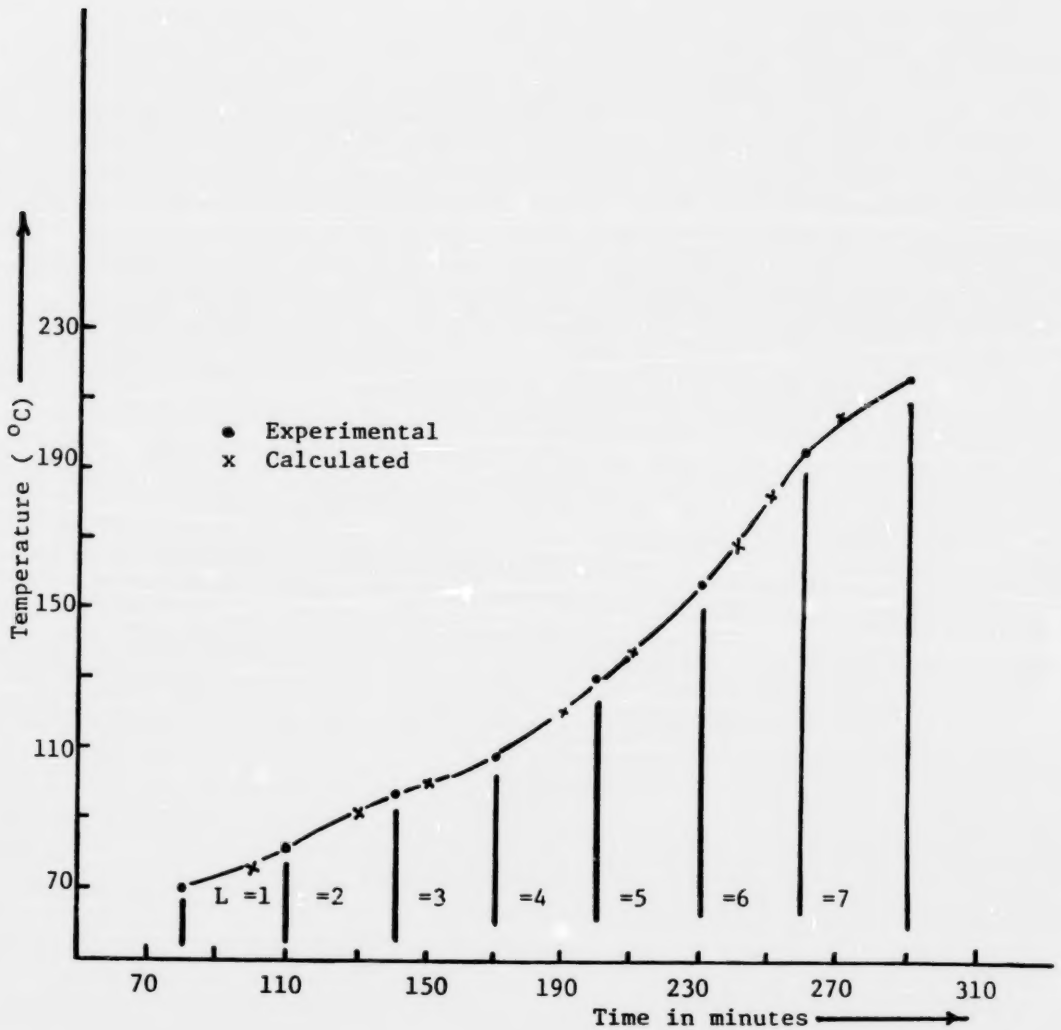

Fig. 2 Measured and interpolated temperature using cubic spline formulation(coal A).

Table 1

1) Measured and calculated temperature and 2) calculated change and rate of change of temperature with respect to time for the high volatile bituminous coal A.

L	MEASURED TEMPERATURE °C	CALCULATED TEMPERATURE °C	CHANGE OF TEMPERATURE WITH TIME °C/min.	RATE OF CHANGE OF TEMPERATURE WITH TIME
1	81.17	81.17	0.5308	0.0045
2	96.40	96.40	0.3938	
3	107.48	107.48	0.5250	0.0224
4	129.60	129.60	0.8262	
5	157.17	157.17	1.1392	0.0232
6	195.39	195.39	1.1959	

Table 2

Bicubic spline coefficients for the high volatile bituminous coal A for six intervals (L=1,2,...,6).

	CUBIC SPLINE COEFFICIENTS		
L	C(L,1) α(L,1)	C(L,2) α(L,2)	C(L,3) α(L,3)
1	0.1231424	0.0113318	-0.0001008
2	0.5307620	0.0022556	-0.0001008
3	0.3938097	-0.0068206	0.0002002
4	0.5249996	0.0111936	-0.0001372
5	0.8261924	-0.0011539	0.0001416
6	1.1392306	0.0115885	-0.0002365

TABLE 3.

1) Measured and calculated temperature; and 2) calculated change and rate of change of temperature with respect to time for the low volatile bituminous coal B.

L	MEASURED TEMPERATURE °C	CALCULATED TEMPERATURE °C	CHANGE OF TEMPERATURE WITH TIME °C/min.	RATE OF CHANGE OF TEMPERATURE WITH TIME
1	93.16	93.16	0.5628	0.0014
2	110.00	110.00	0.5375	
3	125.65	125.65	0.5360	0.0030
4	142.80	142.80	0.5985	0.0012
5	161.67	161.67	0.6721	0.0037
6	182.73	182.73	0.7062	

TABLE 4

Bicubic spline coefficients for the low volatile Bituminous coal B for six intervals (L=1,2,...,6).

	CUBIC SPLINE COEFFICIENTS		
L	C(L,1) α(L,1)	C(L,2) α(L,2)	C(L,3) α(L,3)
1	0.4542169	0.0029253	-0.0000248
2	0.5628083	0.0006944	-0.0000248
3	0.5375499	-0.0015364	0.0000336
4	0.5359919	0.0014844	-0.0000098
5	0.5984830	0.0005986	0.0000140
6	0.6720761	0.0018545	-0.0000286

REFERENCES

1. Chauvin, R., Measurements of the Reactivity of the Solid Fuels by the Crossing-point Method. Rev. Gen. Thurm, 3 (26), 1964, p.159.

2. Coward, H.F., Research on Spontaneous Combustion of Coal in Mines - A Reviw, SMRE RR-142, 1957.

3. Feng, K.K., Chakravorty, R.N., Cochrane, T.S., Spontaneous Combustion-A Coal Mining Hazard, 7th Annual General Meeting of the CIM, Vancouver, April 1973, pp. 1-10.

4. Chakravorty, R.N., Kar, K., Characterization of Western Canadian Coals with Respect to Their Susceptibility to Spontaneous Combustion, Coal Reseach Laboratories, CANMET, Div. Report, ERP/CRL 86-151, December 1986.

5. Ahlberg, J.H., Nilson, J.L., Walsh, J.L., The Theory of Splines and Their Applications, New York: Academic Press, 1967.